

EFFECT OF DREDGING ON HG DISTRIBUTION IN WATER AND SEDIMENTS IN THE MUCURIPE HARBOR, FORTALEZA, NE, BRAZIL

Efeito da dragagem na distribuição de Hg em água e sedimentos no Porto Mucuripe, Fortaleza, NE, Brasil

Luiz Drude de Lacerda¹, Ingra Kellen Cavalcante Belmino², Lucas Moreira Buruaem³, Rozane Valente Marins⁴

¹ Professor do Labomar/UFC e pesquisador do CNPq

ABSTRACT

Dredging and tailings disposal can mobilize mercury (Hg) accumulated in bottom sediments, increasing the exposure of the aquatic biota. In the Mucuripe Harbor, NE, Brazil, dredging is performed regularly, but the impact on Hg mobilization is unknown. This paper presents results from the monitoring of a dredging operation to characterize and quantify an eventual Hg mobilization. The results showed that remobilization is significant and is associated with suspended solids. Further deposition of the Hg-enriched, remobilized, suspended solids increased Hg concentrations by a factor of 1.2 and 2.0 in harbor and shelf sediments, respectively. Maximum concentrations in harbor and shelf sediments reached 79.9 ng.g-1 and 32.4 ng.g-1 (20 and 9 higher than the regional background, respectively). The progressive increase in Hg concentrations may result in higher exposure to the local biota.

Keywords: metals, mobilization, contamination, coastal sediments.

RESUMO

Dragagem e destinação de rejeitos no porto do Mucuripe, em Fortaleza, NE, Brasil, aumentaram a mobilização de Hg associada aos sólidos suspensos e às concentrações de Hg por um fator de 1,2 e 2,0 em sedimentos de porto e plataforma continental, respectivamente.

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² Cientista ambiental e mestra em Ecologia pela UFC

³ Biólogo e pesquisador do Núcleo de Estudos em Poluição e Ecotoxicologia Aquática (Nepea), Unesp, São Vicente, SP ⁴ Professora do Labomar/UFC, coordenadora do Laboratório de Biogeoquímica Costeira e pesquisadora do CNPq E-mail par correspondência: Idrude1956@gmail.com

As concentrações máximas em sedimentos do porto, de 79,9 ng.g-1, embora baixas quando comparadas a outras áreas contaminadas em portos brasileiros, representam um aumento de 20 vezes relativo ao valor de fundo regional. Os sedimentos adjacentes da plataforma continental, com concentrações máximas de 32,4 ng.g-1, também influenciados por rejeitos, são 9 vezes maiores. O aumento progressivo das concentrações de Hg pode estar associado ao teor de Hg anormalmente elevado observado em peixes locais.

Palavras-chave: metais, mobilização, contaminação, sedimentos costeiros.

INTRODUCTION

Dredging to keep navigable harbor accesses and maneuvering areas changes the chemical and biological characteristics of bottom sediments and the overlying water column, both at the operation and at disposals sites (Caplat *et al.*, 2005), generally increasing sediment toxicity (Baruaem *et al.*, 2012; Moreira *et al.*, 2017; Saes *et al.*, 2019). Amongst these changes, the mobilization of contaminants is of relevant environmental concern, in particular those elements quickly incorporated into food chains that may result in increasing exposure to pollutants. The importance of such an environmental issue led to the promulgation of a specific regulatory legislation in many countries. In Brazil, the Conama Resolution 344/2012 (Conama, 2012) establishes legal limits for pollutant concentrations in sediments subjected to dredges and their disposal. However, results from different harbor areas worldwide, strongly suggest that even with concentrations lower than legal limits increasing exposure to pollutants may cause deleterious effects to the biota (Van den Hurk *et al.*, 1997).

Among the contaminants which can suffer chemical changes due to dredging, Hg has been reported to suffer intense remobilization, having its chemical speciation in water changed, its distribution in bottom sediments modified and its bioavailability to the biota increased, due to the mobilization of readily bioaccumulated species, in particular methyl mercury (MeHg). Mercury is regulated under the Conama (2012) act and the proposed limit concentrations in sediments range from the Level 1 (300 ng.g⁻¹), when no specific disposal is required, to Level 2 (1,000 ng.g⁻¹), when pre-disposal treatment is required. These concentrations, however, may be 10 to 100 times higher than the local background of Hg in Brazilian shelf sediments (Marins *et al.*, 2004). A critical review of Hg sediment quality values (SQV) for the protection of benthic invertebrates shows low predictive value of SQVs when Hg concentrations are above background levels (Conder *et al.*, 2015), in Brazilian shelf sediments background Hg concentrations are on the order of 5 to 30 ng.g⁻¹ (Marins *et al.*, 2004). Therefore, virtually any dredging of merely Hg-enriched sediments can pose environmental threats to aquatic organism.

The top few centimeters of the sediment is the most dynamic layer, both for Hg methylation and diagenesis, therefore, even small dredging operations can increase Hg bioavalability (Bloom & Lasorca, 1999). In addition, during dredging and disposal of tailings, Hg present in sediments reacts with other compounds such as organic carbon and sulfate in an oxic environment maximizing its remobilization (Shultz *et al.*, 1995; Aquavita *et al.*, 2012). Once mobilized, Hg can enter food chains increasing the environmental exposure to the biota and humans, through the ingestion of contaminated fish, in particular.

In the Mucuripe Harbor, the oldest and largest maritime terminal in Fortaleza, NE Brazil, navigation canals and maneuvering areas require maintenance dreading on a 3 to 5 year basis. In the last dredging in 2011, the navigation canal was deepened to 14 m and the activity removed about 5.7 million cubic meters of sediments that were discharged offshore. During this period a monitoring program was established to follow eventual changes in the hydrochemistry, the distribution and composition of the local biota and the presence of contaminants (CDC, 2010). Since in the Mucuripe coastal region, previous studies on Hg distribution in the local environment had reported abnormal Hg concentrations in sediments (Marins et al., 2004; Baruarem et al., 2012) and in fishes commercialized in the local market (Costa & Lacerda, 2009), the dredging and tailings disposal may contribute to a further increase in Hg bioavailability. Unfortunately, the impact on these activities on the Hg mobilization in the Mucuripe Harbor area is still unknown. Therefore, the accompanying of Hg concentrations in water, suspended particles and bottom sediments, in harbor and the adjacent shelf area during the 2011 dredging were performed, in order to verify if any Hg mobilization occurs and the mechanism responsible for any eventual increasing in Hg concentrations.

MATERIAL AND METHODS

In the harbor and in adjacent areas, sampling stations were established as depicted in Figure 1 for water and sediment collection. For the Hg quantification in water (9 stations), duplicate samples we collected only during the dredging operations, since any changes in Hg concentrations in water remain for only a short period previous to dilution and/or incorporation onto the solid phase (Salomons & Forstner, 1984). Sediment samples (10 stations) were collected before (2007) and after the dredging, in 2011. Four (4) additional

sampling stations for the collection of shelf sediment were established in areas outside the harbor, sampling only occurred once in 2011. These additional stations were selected to coincide with previously sampled stations in 2002 (Maia, 2004) and were used for comparative purposes of temporal trends in Hg concentrations in local sediments.

Duplicate water samples were collected in Teflon 500 ml bottles and immediately filtered in the laboratory (0.45μ) to determine dissolved Hg (Hg-D). Filtered water samples (40 ml) were digested with

Atlantic Ocean

Sediment samples

Was as 28' W

Atlantic Ocean

Sediment samples

Was as 28' W

Atlantic Ocean

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Atlantic Ocean

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Atlantic Ocean

Fortaleza

Figure 1 – Stations grid of sediment and water samples in the Mucuripe Harbor and adjacent shelf sediments, off Fortaleza, Northeastern Brazil

38° 30' W

38° 28' W

7.5 ml de HCl (4N), and 1.0 ml of a KBrO $_3$ /KBr (0.1N) solution in Boeco glass vials for 30 min; after, 25 µL of hydroxylamine (12%), were added to reduce the excess potassium bromine. Particulate Hg (Hg-P) were measured after the digestion of filters with 20 ml of a 50% aqua regia solution (H $_2$ O:HCl:HNO $_3$; 4:3:1) in 125 ml Erlenmeyer flasks at 70 °C for one hour (Marins *et al.*, 2002a). Hg-D and Hg-P were quantified in a PSA Millennium Merlin 10.025 cold vapor atomic fluorescence spectrometer (CVAFS), after reduction with SnCl $_2$. The analytical precision, estimated as the relative percent difference between aliquots, was always less than 20%. The recovery of known additions of Hg $^{2+}$ to filtered marine waters averaged 92 \pm 4%.

Duplicate sediment samples were collected using a Van Veen dredge, packed in plastic bags and kept refrigerated till analysis. Subsamples were dried at 50 °C till constant weight and sieved through 0.063 mm mesh. Dried duplicate samples weighting 1.0 g were leached with 20 ml of 0.1 N HCl after 2 hours contact in a shaker to extract the exchangeable fraction (Hg-Exch). Sequentially samples were digested in with 20 ml of a 50% aqua regia solution (H₂O:HCl:HNO₃; 4:3:1) in 125 ml Erlenmeyer flasks at 70 °C for two hour to extract the strongly bound Hg fraction (Hg-Sb) present in sediments (Aguiar *et al.*, 2007). Hg-Exch and Hg-Sb were quantified in a Nippon Instruments Corporation (NIC) model RA3210A cold vapor atomic absorption spectrometer (CVAAS), after reduction with SnCl₂. For quality assurance of the Hg determination in the TSS and sediments, a certified reference material (NRC PACS-2, Canada) was simultaneously analyzed (n = 6), and the analysis demonstrated an average Hg recovery of 103 ± 4%.

The Hg detection limits, estimated as three times the standard deviation of reagent blanks, was 0.30 ng.g⁻¹ in suspended matter and sediment samples and 0.18 ng.l⁻¹ in water samples. In all cases, the blank signals were lower than 0.5% of sample analysis. The concentration values were not corrected for the relative recoveries obtained for the certified material.

RESULTS AND DISCUSSION

Average Hg-D concentrations were below the detection limit of the method in all samples (< 0.18 ng.l⁻¹), confirming that if any alteration on dissolved Hg occurs they are short-lived or involve very low concentrations. Dissolved Hg concentrations in coastal waters are generally lower than the detections limit of this study. Marins *et al.* (2002b) reported dissolved Hg concentrations in the contaminated Ceará River lower estuary between 1.2 and 1.7 ng.l⁻¹ and of 0.06 to 1.07 ng.l⁻¹ in the Pristine Pacoti river lower estuary. Shelf waters present even lower concentrations varying from < 0.05 to 0.48 ng.l⁻¹ (Lacerda & Marcovecchio, 2018). Thus the relatively high detection limit and the single sampling strategy hamper a discussion of a possible impact of the dredging operations on the dissolved Hg concentrations.

Mean and range of concentrations of Hg-P in water samples collected inside and outside the harbor area showed no significant difference, similar to the suspended particulate matter content (TSS). However, the Hg concentration in the TSS was significantly (P < 0.05) higher inside the harbor area than in the outside samples (Table I), strongly suggesting a change of quality of the TSS through the contribution of Hg-enriched dredged materials to the suspended matter inside the harbor area. Typical concentrations in the TSS of coastal areas of Ceará state vary depending on Hg inputs, but in areas receiving effluents

from the Fortaleza metropolitan region, Hg concentrations range from 13 to 166 ng.g⁻¹ (Marins *et al.*, 2002b).

Enrichment of the TSS with Hg has been reported elsewhere and results either from remobilizing deeper, Hg-enriched sediment layers, and release of dissolved Hg from interstitial waters that readily adsorbs onto the TSS (Bloom & Lasorca, 1999). For example, highest Hg concentrations observed in Sepetiba Bay suspended matter, were suggested a result from frequent dredging for harbor maintenance and reflect ressuspension of bottom sediments, and typically present Hg contents ranging from 80 to 140 ng.g⁻¹ (Lacerda *et al.*, 2001).

Table I – Suspended particulate matter, Hg-D and Hg-P in waters sampled inside (n = 10) and outside (n = 8) the harbor area during the 2011 dredging in the Mucuripe Harbor region, Fortaleza, NE, Brazil

Area	TSS (mg. 1 ⁻¹)	Hg-D (ng.1 ⁻¹)	Hg-P (ng.l ⁻¹)	Hg-TSS (ng.g ⁻¹)
Outside	21.2 ± 1.8 19.0 - 22.6	< 0.18 ng.l ⁻¹	2.10 ± 0.38 1.75 - 2.54	99.0 ± 16.5 77.4 - 112.3
Inside	19.2 ± 3.2 15.6 – 22.2	< 0.18 ng.1 ⁻¹	2.45 ± 0.17 2.32 - 2.41	129.7 ± 17.3 111.5 - 151.5

Sediment Hg concentrations, TOC and fine sediment contents measured before and after the dredging operation are shown in Table II. Data from 2007, prior to the dredging activities, showed harbor sediments enriched in Hg, TOC and fine sediments, relative to shelf sediments outside the harbor area. After the dredging, which deepened harbor depth from 5-6 to 10-13 m, both harbor and shelf sediments were enriched in Hg and fine sediments. Regarding the TOC content only shelf sediments were enriched after the dredging. Hg concentrations increased by 1.2 and 2.0 times in harbor and shelf sediments, respectively, from 2007 to 2011. However, the increase in Hg concentrations was more significant in shelf sediments. In 2007 harbor/shelf Hg concentrations ratio was 3.0 (34.6 to 11.5 ng.g⁻¹), and decreased to 1.8 (41.6 to 22.8 ng.g⁻¹) in 2011 after the dredging, meaning that concentrations in shelf sediments were nearly doubled due to tailings disposal. Fine sediment contents increased 6.1 and 8.3 times in harbor and shelf sediments, respectively, from 2007 to 2011. The higher concentrations of Hg and fine sediments in the harbor are after the dredging, suggest that exposure of deeper layers occurred, and is consistent with high Hg concentrations observed in the TSS.

Maximum (79.9 ng.g⁻¹ d.w.) and average (41.6 ng.g⁻¹ d.w.) Hg concentrations in harbor sediments are low compared to other harbor areas in Brazil. In sediments of the largest Santos Harbor in SE Brazil, Baruarem *et al.* (2012) reported a maximum (640 ng.g⁻¹ d.w.) and average (260 ng.g⁻¹ d.w.) Hg concentrations; respectively 8.0 and 6.5 times higher than the concentrations observed in the Mucuripe Harbor. Compared to industrial harbors, such as Baltimore and Boston Harbors, the observed concentrations in the Mucuripe Harbor sediments are 10 to 100 times lower (Mason and Lawrence, 1999).

Table II – Mercury, total organic carbon (TOC) and fine sediment ($< 63\mu$) contents is sediments inside the Mucuripe Harbor and in adjacent shelf areas affected by the release of dredge tailings, before (2007) and after (2011) dredging operations. TOC and fine sediment data from Baruaem *et al.* (2012)

Hg (ng.g ⁻¹ d.w.)		TOC (%)		θ < 63μ			
Shelf	Harbor	Shelf	Harbor	Shelf	Harbor		
2007							
11.5 ± 13.1	34.6 ± 8.2	0.4 ± 0.4	1.1 ± 0.3	6.5 ± 8.7	12.5 ± 10.1		
0.02 - 32.6	25.8 - 46.2	0.1 - 1.1	0.8 - 1.4	0.1 - 16.7	2.4 - 22.7		
2011							
22.8 ± 15.7	41.6 ± 23.9	2.7 ± 5.2	1.0 ± 0.6	54.0 ±	76.3 ± 14.2		
4.4 20.2	24.4 70.0	01 105	0.4 1.0	40.2	F(0 00 1		
4.4 – 38.2	24.4 – 79.9	0.1 – 10.5	0.4 – 1.9	1.8 - 95.3	56.0 – 90.1		

Mean Hg concentrations in shelf sediment reported in 2002 were 3.4 ± 3.0 ng.g⁻¹ (Maia, 2004). In 2011, the same stations doubled their average Hg concentrations to 7.3 ± 1.9 ng.g⁻¹. This confirms a significant increase during this 7-year period. After that sampling, in 2002, the harbor suffered its first two large dredging operations of about 1.0 and 3.0 million cubic meters, in 2003 and 2004, respectively. From its construction in 1991 and 2003, two small operations (< 0.8 million cubic meters) occurred. Therefore, the increased concentrations verified in shelf sediments, may be associated with the dredging operations between 2002 and 2011. All operations discharged tailings in nearby shelf areas.

Moreover, considering the regional background estimated by Marins *et al.* (2004), the average and maximum Hg concentrations in the Mucuripe Harbor sediments (41.6 and 79.9 ng.g⁻¹, respectively), represents a 8 and 20 times increase relative to regional background, whereas the average and maximum concentrations in adjacent shelf sediments influenced by tailings (22.8 and 38.2 ng.g⁻¹, respectively) are 4.5 and 7.5 times higher than the background.

Modeling of the expected increase in Hg uptake by fish, in areas suffering Hg mobilization due to dredging and sediments disposal estimated a 30% increase of Hg concentrations in carnivorous fish (Schultz *et al.*, 1995). At the Kaohsiung Harbor, Taiwan, MeHg level showed a positive correlation with the total Hg and total organic carbon in the sediments, reaching from < 0.01 to 2.66% of the total Hg content in the sediments, suggesting that increasing Hg and TOC contents in sediments, as observed for the shelf sediments at the Mucuripe area, also increase Hg bioavailability (Chen *et al.*, 2018). Similarly, in Baltimore Harbor sediments, Mason and Lawrence (1999) highlighted the importance of organic matter in regions removed from point source input in controlling both the concentration and bioavailability of MeHg to organisms. Based on these and other evidence from dredging of harbor sediments worldwide, as reviewed by Conder *et al.* (2015), the progressive increase in Hg concentrations, together with TOC in sediments of shelf areas surrounding the Mucuripe Harbor may increase the already observed anomalously high Hg content observed in local fish (Costa & Lacerda, 2009; Lacerda *et al.*, 2016).

CONCLUSIONS

Mercury mobilization due to dredging in different parts of the world suggests a significant increase of Hg bioavailability by favoring MeHg production and thus increasing

Hg concentrations in carnivorous fish. These observations prevented or altered sediment dredging and dredging techniques in many regions. Unfortunately, for the Mucuripe Harbor, the scarce data on Hg distribution in the local food chains, as well as of regular monitoring of environmental parameters of the harbor area, impede a cause-effect relationship to be quantified. However, the increasing trend verified in local coastal sediments, between 2003 and 2011, and the Hg enrichment in suspended solids and surface bottom sediments of adjacent areas observed in this study, associated with an enrichment of TOC in these same sediments, may support an association with the Hg concentrations in local fish and of the estimated exposure to Hg by consumers. Therefore, our results strongly suggest the characterization of the Hg behavior and fate should be included in EIA studies of future dredging operation in the Mucuripe Harbor.

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